

Ultrafast Measurements of Liquid Carbon and Liquid Silicon

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INTRODUCTION

Conventional techniques of x-ray absorption spectroscopy, while extremely useful for studying the electronic and atomic structures of many materials, are generally not suitable for high temperature, volatile materials. The introduction of time resolution overcomes much of this difficulty: by melting thin foils with an ultrafast laser pulse and measuring the transmission of x-rays before break-up of the liquid, we can obtain useful absorption spectra of high-temperature liquids.

EXPERIMENT AND RESULTS

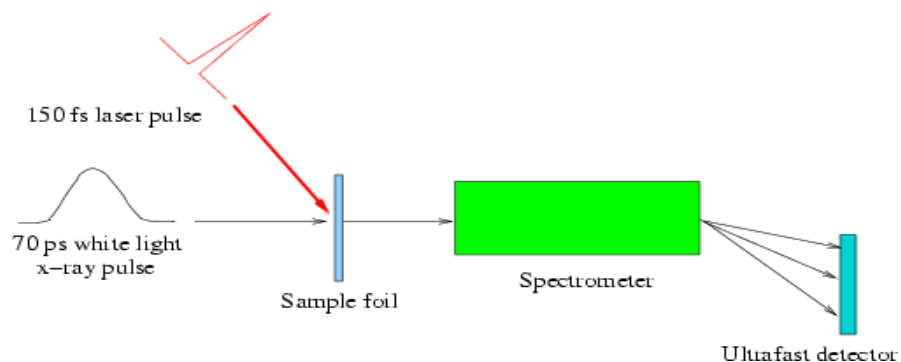


Figure 1. Sketch of experimental setup for time-resolved absorption.

Figure 1 shows a sketch of the experimental setup at beamline 5.3.1 of the Advanced Light Source. After heating the foil with an 800 nm, 150 femtosecond laser pulse, a 70 ps pulse of broad spectrum x-rays from a synchrotron bend magnet pass through the laser-excited region of the foil. A spectrometer then disperses the x-rays onto a detector. The detector is either a set of microchannel plates (for pump-probe measurements with 70 ps time resolution) or an ultrafast x-ray streak camera (for a resolution of up to 1 ps).

Figure 2 compares the L -edge spectrum of an unheated silicon foil with the spectrum of the foil 100 ps after laser excitation. The transition from solid to liquid results in a number of changes to the spectrum: a 50% drop and 2 eV broadening in the $L_{II,III}$ edge at 100 eV, a -1.6 ± 0.2 eV shift in the L_I edge at 150 eV, and a dramatic decrease in the magnitude of EXAFS oscillations. Model calculations based on molecular dynamics simulations of liquid silicon compare favorably with these results.

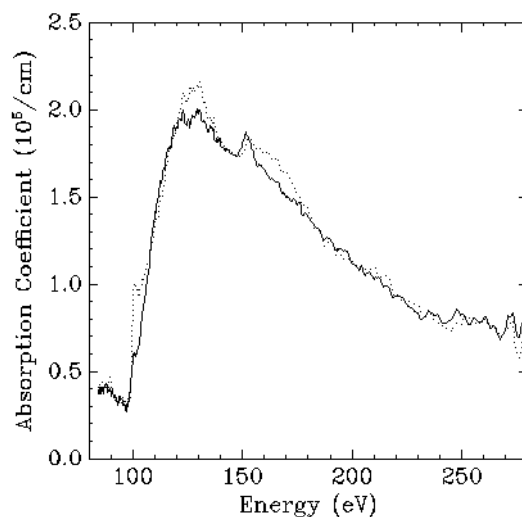


Figure 2. Absorption Spectra of silicon L-edges. Dotted line: unheated silicon; Solid line: heated silicon, 100 ps after laser.

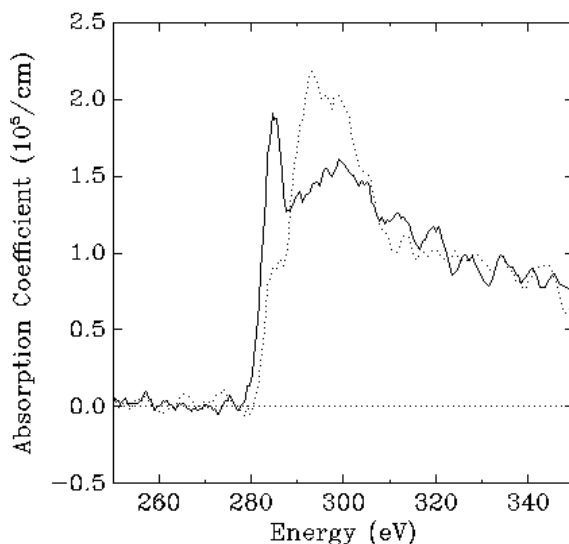


Figure 3. Carbon K-edge absorption spectra. Dotted line: unheated carbon; solid line: heated carbon, 100 ps after laser.

Measurements on foils of soft amorphous carbon, summarized in figure 3, show large differences between the unheated and heated spectra of the carbon *K*-edge. After 100 ps, the laser-induced melting causes an increase in the size of the π^* resonance at 285 eV accompanied by a decrease and 4 eV shift in the σ^* resonance at 296 eV. These changes indicate a transition in bonding geometry from the predominant sp^2 bonding characteristic of soft amorphous carbon to the collection of distorted sp bonded chains recently predicted by molecular dynamics simulations for the low-density liquid phase of carbon.¹

REFERENCES

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